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Hyperfine splitting of the $2s_{1/2}$ and $2p_{1/2}$ levels in Li- and Be-like ions of the open-nuclear shell isotope $^{141}_{59}$Pr

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The isotope $^{141}_{59}$Pr has a closed neutron shell and a wide open proton shell. The hyperfine splittings of the $2s_{1/2}$ and $2p_{1/2}$ levels in Li- and Be-like ions of this nucleus have been measured at a high-energy electron beam ion trap by high-resolution spectroscopy of the $2s_{1/2}$ - $2p_{1/2}$ transitions in the extreme ultraviolet. The results are 191.6 ± 2 meV for the $2s_{1/2}$ ground level and 68 ± 2 meV for the excited level, $2p_{1/2}$, in the Li-like ion, and of 245.8 ± 3 meV for the $2p_{1/2}$ $F = 3/2 - 7/2$ hyperfine interval in the Be-like ion.

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For more than 80 years, the hyperfine splitting of lines in atomic spectra has been studied in the quest for information on nuclear properties. The most sensitive probe is provided by s electrons which are the only ones whose wave function does not vanish at the center of the nucleus. All of the early measurements of hyperfine structure (HFS) employed neutral atoms. However, as HFS effects grow with the third power of the nuclear charge Z, heavy elements are preferred for detailed studies that exploit HFS for learning about nuclear effects, such as the effective nuclear size, the nuclear charge distribution, and the nuclear magnetization distribution, as well as for investigating parity violation (PNC) in order to test the Standard Model. Any interpretation of the HFS in neutral atoms or near-neutral ions requires a thorough understanding of the multi-particle system that forms the electron shells around the nucleus, and this complexity limits the achievable accuracy in extracting nuclear information.

The physics related to the HFS can be disentangled from the complex atomic physics plaguing heavy neutral atoms or near-neutral ions by probing highly charged, heavy ions that feature only a few electrons so that their shell structure is amenable to accurate calculations. Measurements of the hyperfine structure in such highly charged heavy ions have become possible only fairly recently. Of the two successful approaches world-wide, one technique employed the ESR heavy-ion storage ring at GSI Darmstadt, Germany, and sought to induce a laser resonance [1, 2] between the hyperfine levels. Owing to the limited predictive power of theory, especially in the absence of a sufficiently good nuclear model, the search for the resonance was tedious and covered only one isotope at a time. The experiments have produced data for one-electron ions of two isotopes, ^{207}Pb and ^{209}Bi, both of which are near to the doubly magic nucleus ^{208}Pb with its closed proton and neutron shells. No data have yet been produced for few-electron ions. Because ions in a storage ring move at a large fraction of the speed of light, the attainable precision is limited by the precision with which the ion velocity could be determined.

The other technique employed the Livermore SuperEBIT high-energy electron beam ion trap and passive multichannel spectroscopy [3–5]. Since the ion cloud in the trap is practically at rest, there is no Doppler shift problem for the calibration. The attainable precision in this case is typically limited by the spectrometer resolution and the counting statistics. With position sensitive detection, several isotopes can be studied simultaneously. This makes for more data (the data on six of seven isotopes worldwide have been obtained at Livermore), permitting the study of differential effects, such as the variation of the nuclear magnetization distribution in ^{203,205}Tl^{80+} [5], which demonstrated that even close to the doubly magic nucleus $^{208}_{82}$Pb the nuclear spin can no longer be described as a single unpaired nucleon orbiting a core of paired nucleons. A range of models (for example, [6–15]) have since been devised that involve collective or dynamical effects to explain the magnetic field generation in the nucleus. The models differ, for example, in the assessment of the role of the negative energy continuum [10], in the inclusion of dynamic correlations among clusters of nuclear particles, or in what is dubbed the "boiling" of the quantum electrodynamical (QED) vacuum [9, 13].

The SuperEBIT experiments have also been the only ones producing information on the HFS in few-electron ions. These measurements focused on the Li-like (three-electron) ion of ^{209}Bi [16, 17]. The storage-ring method has only recently, after about a decade of trying, produced a first signal [18], and the data analysis is still awaiting the resolution of calibration issues. The study of the HFS of few-electron ions not only provides another stringent test of atomic and nuclear theory, but together with HFS measurements of H-like ions, it enables

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the isolation and testing of QED effects in the strongest magnetic fields accessible on Earth [12].

Here we present the first measurements of the HFS of few-electron ions with nuclei far from closed shells. The measurement focuses on the isotope $^{141}_{59}$Pr (nuclear spin $I = 5/2$). ^{141}Pr with its 82 neutrons has a closed neutron shell, but a proton number (59) that is far away from the nearest magic number value (50). An observation of the $F = 2 - 3$ transition in the ground state of the Li-like ion would require resonance measurements near 5 μm, which at present is not possible to do; present-day storage rings are unable to Doppler-shift the transition into a more friendly wavelength regime. We overcome this limitation by observing the $2s_{1/2} - 2p_{1/2}$ transition, which is in the EUV, using a very-high resolution spectrometer. Such a measurement has the added advantage that we can observe not only the splitting of the $2s_{1/2}$ ground level but also that of the $2p_{1/2}$ excited level. Furthermore, we can observe this splitting in the Be-like ion. The hyperfine-induced decay of the lowest-lying excited $J = 0$ level in Be-like ions has recently found interest because of new experimental lifetime data [19, 20] and the search for an adequate theoretical description [21]. We observe the hyperfine splitting of the nearby $J = 1$ level of the same electron configuration and term, $2s2p\ ^3P^o$, which is responsible for the decay of the $J = 0$ level through hyperfine quenching.

The experiment was done at the Lawrence Livermore National Laboratory SuperEBIT high-energy electron beam ion trap [22]. For the benefit of a high signal rate, most of the present work was performed at electron beam energies of 102 to 108 keV, that is far beyond the ionization potential of Pr^{56+} at 11.9 keV [23], and at electron beam currents up to about 155 mA. Injection of Pr into the ion trap was achieved by means of a plume of material released by a pulsed Nd:YAG laser [24]; the neutral particles that drift across the trap volume are collisionally ionized as they interact with the electron beam and are consecutively ionized further. After about 77 s, the stored ions were dumped by lowering one of the trapping voltages and then a new cycle started. A low-density flux of a light-atom gas (N_2 or trimethylborate (TMB)) provided cooling for the high-charge ions in the trap. At higher injection pressures and with faster trap cycles (because the light ions rapidly lose all electrons), the cooling gas also provided lines for wavelength calibration. The charge state distribution in the trapped ion cloud was monitored using the XRS X-ray spectrometer micro-calorimeter built by NASA/Goddard Space Flight Center [25].

For Li-like ^{141}Pr, the $2s$ $F = 2 - 3$ hyperfine splitting amounts to about 1/500 of the roughly 130 eV $2s - 2p_{1/2}$ transition energy. Observation of such a small effect requires a very-high resolution EUV spectrometer. We used a flat-field grazing incidence spectrometer based on a gold-coated variable line spacing grating of $R = 44.3$ m radius of curvature and 2400 ℓ/mm average groove density set up at an angle of incidence of about 88° [26]. This

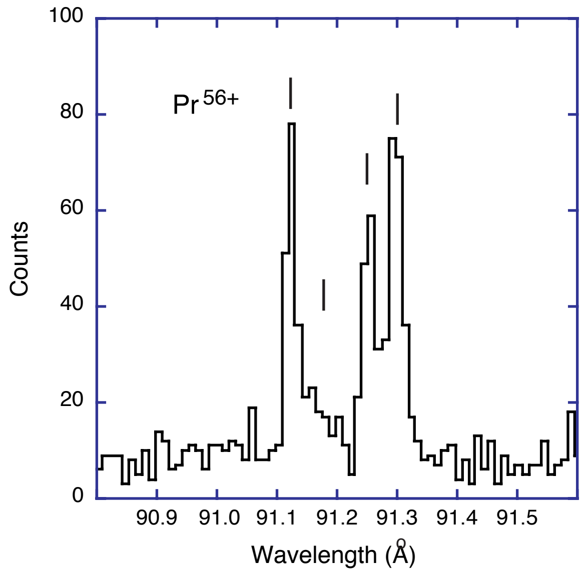

FIG. 1: One of seven sum spectra showing the vicinity of the four lines of the $2s_{1/2} - 2p_{1/2}$ transition array of Pr^{56+}.

spectrometer was equipped with a detector that comprises a cryogenically cooled, thinned, back-illuminated charge-coupled device (CCD) of 1340 × 1300 pixels on a 25 mm × 25 mm substrate. The wavelength coverage is about 15 Å in our range of interest. Each pixel then corresponds to a wavelength interval of about 12 mÅ. As we image the ∼60 μm diameter electron beam with about unity magnification onto the detector, the typical line width (FWHM) is about three pixels, or 35 mÅ, corresponding to a resolving power $\lambda/\Delta\lambda$ of about 2700. Exposure times of individual spectra were 30 minutes or 60 minutes, and 29 such spectra were individually filtered against cosmic radiation hits and then summed in seven independent data sets. Data analysis included fits to each of the data sets and measures of reproducibility (scatter) among the sets.

The spectrometer wavelength scale and dispersion were obtained by calibration with lines of few-electron ions of B (injected as TMB), O (from earlier uses of CO_2), and Ne. In the Pr spectra, a weak third-order line of N VI served as an additional anchor. Accurate reference data for the lines of B, N, O, and Ne are available from several sources [27–30]. One of the seven summed spectra displaying the four hyperfine components of the $2s_{1/2} - 2p_{1/2}$ transition array in Li-like Pr^{56+} is shown in Fig. 1. There are four line spacings between the four lines, two of which correspond to the lower ($2s_{1/2}$) and two to the upper ($2p_{1/2}$) level hyperfine splitting. The inferred HFS spacings are presented in Fig. 2, and the average result is given in Table I. The experimental uncertainties are dominated by the counting statistics.

In Fig. 3 we show the spectral region near 85.8 Å that contains the three hyperfine components of the $2s^2\ ^1P_0 - 2s2p\ ^3P^o_1$ transition array of Be-like Pr^{55+} (de-

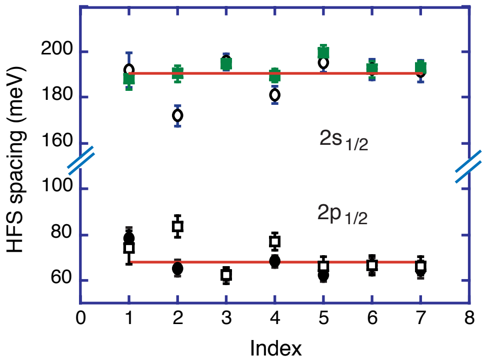

FIG. 2: Spacing of the observed lines from 7 data sets that each comprise the four lines of the $2s_{1/2} - 2p_{1/2}$ transition array of Pr^{56+}. Each data set yields two line spacings (open and closed symbols) for the $2s_{1/2}$ (top) and $2p_{1/2}$ (bottom) intervals. The horizontal lines represent fits to the respective intervals.

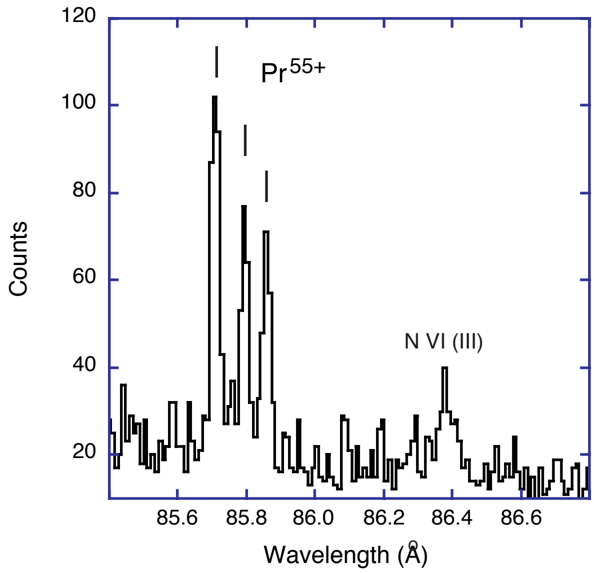

FIG. 3: One of seven sum spectra showing the vicinity of the three lines of the $2s^2\ ^1S_0 - 2s2p\ ^3P_1^o$ transition array of Pr^{55+}. The N VI line appears in third order of diffraction.

TABLE I: Predicted and measured values (in meV) of the hyperfine splitting of the $2s_{1/2}$ and $2p_{1/2}$ levels in $^{141}Pr^{56+}$.

Interval	Source	HFS splitting	Unc.	Ref.
$2s_{1/2}$ $F = 2 - 3$	Experiment	191.6	2.0	This work
	Theory	197.4	0.5	[6]
	Theory	197.48		[10]
	Theory	198.2		This work
[4pt] $2p_{1/2}$ $F = 2 - 3$	Experiment	68	2.0	This work
	Theory	63.62	0.06	[31]
	Theory	63.6		This work

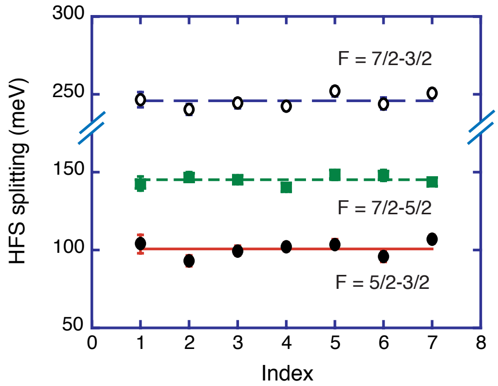

FIG. 4: Hyperfine intervals derived from 7 data sets that each comprise the three lines of the $2s^2\ ^1S_0 - 2s2p\ ^3P_1^o$ transition array of Pr^{55+}, and fits to the data.

cays of the $F = 3/2$, $5/2$, and $7/2$ hyperfine levels to the $F = 5/2$ ground state). The inferred spacings between the three lines are displayed in Fig. 4 and the average for each interval is listed in Table II. For the Be-like ion, the experimental uncertainties are dominated about equally by problems in determining the spectral dispersion (which involves an extrapolation of the wavelength scale) and by the counting statistics.

The results for the hyperfine splittings are compared to theory in Tables I and II. For the $2s_{1/2}$ hyperfine interval in the Li-like ion, our measurement does not overlap within its 1% uncertainty with the calculated values by Shabaev *et al.* [6] and by Boucard and Indelicato [10]. For the $2p_{1/2}$ hyperfine interval in the same ion, the disagreement with the prediction by Korzinin *et al.* [31] is smaller and the difference is only twice the experimental 3% uncertainty. We have also carried out Relativistic Configuration-Interaction (RCI) calculations of these HFS splittings (for the method, see [21]). Our RCI results are close to the other theoretical results and disagree with our measurement. It is interesting to note that while the measured value of the $2s_{1/2}$ $F = 2 - 3$ interval is smaller than predicted, the measured value of the $2p_{1/2}$ $F = 2 - 3$ interval is larger than predicted. A discrepancy between our measured values and theory is also found for the $F = 3/2 - 5/2 - 7/2$ intervals in Be-like $^{141}Pr^{55+}$. The calculations are our own, using the RCI method, as there seems to be no calculation of the hyper-

TABLE II: Predicted and measured values (in meV) of the hyperfine splitting of the $2s2p_{1/2}\ ^3P_1^o$ level in Be-like $^{141}Pr^{55+}$. All results are from this work.

Interval	Source	HFS splitting	Uncertainty
$F = 5/2$ - $7/2$	Experiment	145.0	1.9
	Theory	149	2
$F = 3/2$ - $5/2$	Experiment	100.9	2.2
	Theory	107	2
$F = 3/2$ - $7/2$	Experiment	245.8	3.0
	Theory	256	2

fine splitting in such a four-electron ion in the literature. The measured HFS splittings are consistently and significantly smaller than the calculated values; the deviation amounts to about three times our 1.2% measurement uncertainty for the $F = 3/2 - 7/2$ interval (about twice the combined error bars of experiment and theory).

To gauge the accuracy of our RCI results, we first note that we use a nuclear magnetic moment of 4.2754(5) n.m. for ^{141}Pr from the tabulation of Raghavan [32] that should be accurate enough as to not causing any significant theoretical uncertainties. ^{141}Pr also has an electric quadrupole (E2) moment of -0.0589(42) barn [32], but the E2 contributions to HFS are consistently very small, and at less than 0.1 meV they can be ignored. As for the Bohr-Weisskopf (BW) effect [33], we assume that the nuclear magnetization radius is the same as the nuclear charge radius, which we take to be 6.3154 fm based on the root-mean-square nuclear radius of 4.8919(50) fm from the tabulation of Angeli [34]. Resulting BW corrections are found to be small at about 1 meV, which is also the estimated size of the QED corrections. At this level, these two corrections should not have any significant effects on the accuracy of the calculated HFS splittings. Finally, for high-Z Li- and Be-like ions, electron correlation effects are not expected to be very important. Indeed, even simple multiconfiguration Dirac-Fock calculations give HFS splittings that agree with the highly-correlated RCI results to within a few meV. Overall, uncertainties in the present RCI results are estimated to be around 2 meV, and other theoretical results shown in Table I are likely to be of similar accuracies. That puts theory clearly outside the experimental error bars.

In summary, we have demonstrated that EUV spectroscopy carried out at the high-energy electron-beam ion trap at LLNL is a powerful tool for studying hyperfine structures in high-Z few-electron ions. At present it is not clear what causes the residual difference between theory and experiment. More measurements on other isotopes and ions to provide better comparisons with theory are highly desirable.

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